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TITLE: OVERVIEW OF THE STRATEGIC DEFENSE INITIATIVE PROGRAM IN SHORT WAVELENGTH CHEMICAL LASERS

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OVERVIEW OF THE SDI PROGRAM IN SHORT WAVELENGTH CHEMICAL LASERS

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Advanced chemical lasers promise to be effective space-based weapons against responsive threats. In this program, we are developing both CW and pulsed concepts for achieving this goal. Certain approaches may also be appropriate as ground-based weapons and fusion drivers.

I. INTRODUCTION

Space-based lasers (SBL) continue to be viable candidates for follow-on SDI architectures. Within this category, only space-based chemical lasers (SBCL) and space-based free-electron lasers (SBFEL) are being seriously considered at this time. Engineering maturity of SBCL, resulting from very active development programs over the past 15 years, has thrust this candidate into a leading directed energy weapon (DEW) role in a Milestone II architecture. Other DEW candidates, including ground-based FEL, neutral particle beams, ground-based excimers, and SBFEL, rely upon less mature technology and may be available in future architectures. Additionally, there is a lingering, ever-growing concern that sufficiently intense beams from GBLs cannot be propagated through the atmosphere to perform ballistic missile defense missions.

Current planning for SBCL includes capability growth of these lasers to address responsiveness of the threat. This performance enhancement primarily translates to laser brightness increase, although advances in device weight and expendable weight are obviously important goals, as well.

The baseline SBCL concept incorporates the current Alpha hydrogen fluoride (HF) chemical laser operating at wavelengths near 2.7 μm. Growth in brightness can accrue from larger beam output apertures, from phasing separate apertures, and from lasing wavelength reductions. In this latter category, the nearest-term possibility is the operation on first-overtone transitions within the HF molecule at wavelengths near 1.3 μm. Another possibility, also in this wavelength range, is use of a chemical oxygen-iodine laser (COIL), a device requiring further development for effective space-based use.

There appear to be substantial gains to be made in reducing the wavelength even further. The SDI program charged with accomplishing this goal is the

short-wavelength chemical laser (SWCL) program. In this basic research effort, we are attempting to develop viable chemical lasers operating in the visible and near-visible wavelength range. This paper overviews the SDIO SWCL program.

II. STATUS OF CHEMICAL LASER TECHNOLOGY

Most existing chemical lasers are based upon hydrogen-halide (HX) molecules operating on low-lying, single-quantum vibrational transitions within the electronic ground state of the HX product molecule. The most powerful and useful laser within this category is the HF laser, whose primary reaction step is the "cold" reaction,

$$F + H_2 \rightarrow HF(v) + H \quad . \qquad (1)$$

This fast reaction effectively partitions its exoergicity of ~35 kcal/mole into HF(v = 1, 2, and 3). The F-atoms for the above reaction are efficiently produced in a precombustor upstream of the supersonic mixing region for Reaction (1) by reacting D_2 with (excess) F_2. Three important attributes of the HF laser are the low-mass reactants, reactant availability, and relative simplicity of the kinetics. Favorable fundamentals have driven this laser weapon candidate to a significant engineering maturity. A companion paper[1] describes this status.

COIL, discovered in 1978,[2] is the only chemical laser operating on an electronic transition. In this laser, an energy-storage molecule is generated chemically and then collisionally transfers its energy to the lasing species. The robust energy-storage molecule is created in a solution reaction between gas-phase Cl_2 and liquid H_2O_2. After being formed with near-unity efficiency, the O_2^* (singlet delta at ~1 eV) diffuses through the liquid and flows in the

[1] J. Miller, these proceedings.

[2] W. E. McDermott, N. R. Pc

gas phase to a mixing region where I_2 is injected into the O_2 and diluent. The molecular iodine is first dissociated in collisions with O_2^* after which collisional energy transfer occurs in the fast, near-resonant

$$O_2^* + I \rightarrow O_2 + I^* \quad , \tag{2}$$

The excited atomic iodine is the upper level of the well-characterized and favorable 1.3 μm laser transition.

In addition to its operating wavelength being half that of the HF laser, a very positive feature of COIL is the achievable single-line output spectrum. This property will allow more efficient and simpler multiple-aperture systems. A major drawback to COIL, especially for space deployments, is the cumbersome 2-phase O_2^* generator.

III. EARLY SWCL APPROACHES

Serious efforts directed toward the development of visible-wavelength chemical lasers began in the early 1970s. The emphasis at this time was on identifying chemical reactions of sufficient energy release that electronically excited states in the product species would be energetically allowed. Metal-oxidizer gas-phase reactions dominated the picture.[3] By today's standards, our view was quite primitive, due in large part to the paucity of literature at that time on the participation of electronic states in chemical reactions. At any rate, the generic reaction types were

$$M + RO \rightarrow MO^* + R \quad , \tag{3}$$

and

$$M + RX \rightarrow MX^* + R \quad . \tag{4}$$

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[3] For an excellent review of the status of chemical lasers in the late 1970s, refer to T. A. Cool, "Chemically Pumped Lasers", in Methods of Experimental Physics, Vol. 15B, C. L. Tong, ed., Ac. Press, New York, 1979.

Many of the flames resulting from these very energetic reactions were observed to be very bright, and in several cases, large yields of visible photons were measured. Unfortunately, these photons generally originated from a large number of states. Only modest specificity into electronically excited states was suggested in any of these early studies.

These results are not surprising in that a statistical distribution over product states should be expected in the absence of selection rules. The conservation of electronic spin is thought to be a "good" rule. Reactions (3) and (4) in most cases had no selection rules operating in their favor because the several energetically allowed states, including the ground states, were of the same symmetry. The precondition for the selective production of a specific excited state is that this state must differ in symmetry from the other energetically allowed states. This state will, therefore, be metastable. If it is strongly metastable, i.e., different spin from the ground state, then it is probably characterized by a radiative lifetime too long for a good upper laser level. This excited species, therefore, would be useful as a storage species, similar to O_2^* in COIL. For these reasons, it is now widely accepted that the direct chemical production of a good lasing species is unlikely, except in special circumstances.

IV. LATER SWCL APPROACHES

Beginning in the late 1970s, there were several small efforts in SWCL research. Most of these had turned away from the earlier metal-oxidizer approaches to basically non-metal reactions for which spin-selection rules predicted favorable state specificity. There have been measurements on a few gas-phase chemical reactions showing quite specific production of an excited state. These reactions are shown in Table I.

TABLE I

Reaction	Excited State Energy	Yield	Ref.
$H + NF_2 \rightarrow NF(a) + HF$	1.4 eV	0.90	4
$N + N_3 \rightarrow N_2(A) + N_2$	6.1	$\geq$0.20	5
$O + CN \rightarrow N(^2D) + CO$	2.4	0.85	6

These reactions, along with the one producing O_2^* in COIL, provide positive support for the challenge of SWCL development. However, we must accept the fact that our attempts to increase the excitation energy of the excited species beyond the current 1 eV in the only existing chemically pumped electronic-transition laser lead to greater challenges. It can be argued that COIL is a somewhat special case, primarily having to do with the especially robust character of O_2^*, which, in turn, is due in large part to the low excitation energy of this species.

V. CURRENT SWCL PROGRAM

A. General Approach

The SDIO SWCL program, which began in 1985, attempts to build upon the status prevailing at that time in order to develop a viable laser concept in a timely manner. We are seeking approaches offering high mass efficiency, near-uv or visible wavelength, simple reaction schemes, minimal electrical power requirements, and suitable reactants. The key guidelines are listed in Table II. The ambitious fuel efficiency goal of 1 MJ/kg is for an undiluted stoichiometric flow of the reacting species required in the laser concept.

We envision a supersonic continuous-wave (CW) laser configuration similar to that for the HF laser. A simplified version is shown in Fig. 1, in which the flow is supersonic in the region of the laser axis by virtue of the nozzle expansion. The large excitation energies carried by the electronically excited states offer very high power density possibilities in the flow, which enables relatively compact systems. Using conservative assumptions, Table III shows the expected powers in flows of various excited-state densities through a 10 x 100-cm nozzle exit plane. If moderate densities of a few torr of excited states can be realized, significant SWCL payoffs in system volume and weight can be expected.

The critical technical issues associated with the development of scalable SWCLs are shown in Table IV. Basically, these issues are related to the chemical production of a specific electronically excited state and maintaining it at usable densities over a period of time sufficient to use it. For example, it is undesirable for the excited state to radiate spontaneously at a much faster rate than the reagents can be mixed in the flow.

There are four basic approaches to SWCL development being pursued at this time:

1. direct chemical production of laser species,
2. indirect or storage species/transfer,
3. energy pooling of lower-energy species, and
4. pulsed, premixed schemes.

The first approach, as stated earlier, is expected to be effective only in special circumstances. The second case is preferred because very specific production of excited states can be realized. These states are sufficiently metastable that their energy must be collisionally transferred to be useful. A

subset of this approach is the use of energy pooling, which is attractive because lower-energy species are apparently easier to generate and maintain at high densities. For example, pooling of NF(a) and I* is sufficiently energetic to produce NF(b), which in transition to NF(X) emits green photons. Finally, premixed schemes, by definition, obviate reagent mixing, one of the major SWCL issues. We imagine in this case that a chain reaction will be initiated in an energetic mixture, producing a pulse of excited states and pulsed lasing. Table V shows generic reactions for these four approaches.

Throughout the SDI SWCL program, approximately 15 concepts have been examined. Since space does not permit discussion of each of these, a few specific reaction systems will be overviewed. Further detail can be found in the open literature.

B. Specific Concepts

1. Direct chemical productions of laser species

The $O + N_3$ reaction represents one of several concepts based upon azide chemistry. Many azide reactions appear to be favorable SWCL candidates, based upon the energetic nature of the N_3 radical and the angular momentum constraints that may operate in specific reactions. As discussed earlier, a high degree of specificity is not expected in reactions creating states that are optically connected to the ground state. However, if the energetics of the reaction and the product are such that only a very few product states are energetically possible, then, a statistical distribution over the product states may be sufficient for efficient laser operation. The $O + N_3$ reaction falls in this category because only the X, A, and B doublet states of NO are allowed on energetic grounds. One might expect a substantial fraction of the products to appear in NO(A) and NO(B). A significant yield of NO(A) has been

measured.[7] Other factors that make the NO γ-band laser candidate attractive are its successful lasing in direct optical pumping,[8] low-mass reactants, and availability of the basic reagents. The rate coefficient for $O + N_3 \rightarrow NO + N_2$ has been measured to be 1×10^{-11} $cm^3 sec^{-1}$, and measurements are underway to quantify the yield[7]. Experiments directed toward pulsed production of the reactants in a laser cell are in progress and are expected to lead to a demonstration of a pulsed, chemically driven $NO(A \rightarrow X)$ laser.[7]

2. Storage species/collisional transfer

Chemical production of metastable species has been shown to occur with high specificity in a few instances. The generation of metastable nitrogen molecules by means of the azide reactions $N + N_3$ or $N_3 + N_3$ is believed to be moderately specific. One specific use of this energy is through the reaction sequence

$$N_3 + N_3 \rightarrow N_2(A) + 2N_2 \quad , \tag{5}$$

$$N_2(A) + NO \rightarrow N_2(X) + NO(A) \quad , \tag{6}$$

$$NO(A) + h\nu_L \rightarrow NO(X) + 2h\nu_L \tag{7}$$

In the case of CW operation, the azide radical would be produced by

$$F + HN_3 \rightarrow N_3 + HF \quad . \tag{8}$$

Again, the reagents are lightweight and appear to be readily obtainable. Pulsed demonstration of these kinetic steps is currently being pursued.[7]

3. Energy pooling of lower-energy species

This approach is a subset of the one above but is important enough to be highlighted. High densities of species having excitation energies greater than ~2 eV are difficult because of the greater number of collisional loss channels for higher states. This limitation on the generation of 3-4 eV species can be circumvented by pooling the energy of two long-lived, lower-energy excited states.

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[7] R. D. Coombe, private communication.
[8] M. D. Burrows, S. L. Baughcum, and R. C. Oldenborg, Appl. Phys. Lett. 46, 22 (1985).

We are examining the efficient production of NF(b) by means of:

$$H + NF_2 \rightarrow NF(a) + HF \quad (9)$$

$$NF(a) + HF(v) \rightarrow NF(b) + HF(v-2) \quad (10)$$

$$NF(b) + h\nu_L \rightarrow NF(X) + 2h\nu_L \quad (11)$$

It is possible that this reaction scheme, suggested and researched earlier by Herbelin and Cohen[9] can be effective in existing HF chemical laser devices, such as Alpha.

4. Pulsed, premixed reaction schemes

In order to circumvent the problem of slow fluid mixing, we are addressing the approach of initiating a chain reaction in an energetic mixture of reagents. The initiation energy, either optical or electrical, should be small compared with the chemical energy release in order that the laser system and fuel weight be acceptably low.

A hybrid chemical/excimer laser concept[10] is being examined and has the following azide-based kinetic scheme occurring in premixed XeF_2/HN_3/diluent:

$$h\nu_i + XeF_2 \rightarrow Xe + 2F, \quad (12)$$

$$F + HN_3 \rightarrow HF + N_3 \quad (13)$$

$$N_3 + N_3 \rightarrow N_2^* + 2N_2 \quad (14)$$

$$N_2^* + XeF_2 \rightarrow N_2 + XeF(B) \quad (15)$$

$$XeF(B) + h\nu_L \rightarrow XeF(X) + 2h\nu_L \quad . \quad (16)$$

The reaction sequence is initiated by photolysis of XeF_2. This exciting concept will possibly combine the best features of chemical and excimer lasers. We expect that other reaction schemes within this hybrid category are also possible.

One other example of a pulsed SWCL candidate is one in which the lasing specie is the lead atom.[11] The reaction scheme is initiated by detonating

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[9] J. M. Herbelin and N. Cohen.
[10] R. D. Mead, et al., Proceedings of SPIE Meeting, Conference on Short-Wavelength Lasers, 1988.
[11] I. Bar, et al., ibid.

solid lead azide, after which lasing would occur in the gas-phase expansion zone:

$$Pb(N_3)_2 \xrightarrow{det.} Pb + 2N_3 \quad (17)$$

$$N_3 + N_3 \rightarrow N_2^* + 2N_2 \quad (18)$$

$$N_2^* + Pb \rightarrow Pb^* + N_2 \quad (19)$$

$$Pb^* + h\nu_L \rightarrow Pb + 2h\nu_L \quad . \quad (20)$$

Again, the pump energy is generated by internal chemistry, but in this case, the required pump power density is reduced by virtue of the lasing species being atomic.

A number of research groups from universities, industry, and a national laboratory have contributed to this program. Since all projects could not be specifically mentioned, the past and current participants are listed below:

Ben-Gurion University
Hercules, Inc.
Los Alamos National Laboratory
McDonnell Douglas Research Laboratory
Physical Sciences, Inc.
Rice University
Rocketdyne Division of Rockwell
Rockwell Science Center
Spectra Technologies, Inc.
SRI International
University of Denver
University of South Florida
University of Southern California

VI. CONCLUSIONS

A short-wavelength chemical laser is the Holy Grail[12] of high-performance, high-power lasers. We expect to take a large step in this quest within the next year through our current demonstration projects. A better understanding of SWCL operation and scaling relations should be the immediate outcome from this part of the SWCL program. Meanwhile, the basic research efforts will continue to germinate and nurture new concepts.

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[12] L. C. Marquet, not-so-private communication.

The primary impetus for pursuing this challenge is currently the large payoff expected for the high-brightness, space-laser weapon application. Other SDI missions may also be appropiate. Among these is a space-based illuminator or ground-based weapon. In this latter application, the higher efficiency that may be possible using internal chemistry in pulsed uv or visible laser concepts would be a large step in an SDI ground-based excimer laser improvement program. Finally, and also because of greater possible electrical efficiencies and the consequent reduced cost, pulsed SWCL concepts may one day be appropriate for advanced inertial confinement fusion drivers.

I would like to give a great deal of credit to the many fine researchers who have participated in this SDI research program. This program is sponsored by SDIO/T/IS and is managed by the Los Alamos National Laboratory.

REFERENCES

1. Miller, J.,

2. McDermott, W. E., N. R. Pchelkin, D. J. Bernard, and R. R. Bousek, Appl. Phys. Lett., 32, 469 (1978).

3. Cool, T. A., "Chemically Pumped Lasers", in Methods of Experimental Physics, Vol. 15B, C. L. Toug, ed., Ac. Press, New York (1979).

4. Malins, R. J., and D. W. Setser, J. Phys. Chem, 57, 28 (1981).

5. David, S. J., and R. D. Coombe, J. Phys. Chem., 89, 5206 (1985).

6. Schmatjko, K. J., and J. Wolfrum, Ber. Bunsenges Phys. Chem. 82, 419 (1978).

7. Coombe, R. D., private communication.

8. Burrows, M. D., S. L. Baughcum, and R. C. Oldenborg, Appl. Phys. Lett. 46, 22 (1985).

9. Herbelin, J. M., and N. Cohen, The Chemical Production of Electronically Excited States in the $H+NF_2$ System", Chem Phys. Lett. 20, 603 (1973).

10. Mead, R. D., et al., Proceedings of SPIE Meeting, Conference on Short-Wavelength Lasers, 1988.

11. Bar, I., et al., ibid.

12. Marquet, L. C., not-so-private communication.

- o SPECIFIC STATE PRODUCTION
- o SPECIES PROLIFERATION IN FLOW
- o MAINTENANCE OF HIGH STATE DENSITIES
- o AVAILABILITY OF REACTANTS
- o REACTANT FLOW DENSITIES
- o RADIATIVE LIFETIME/MIXING TIME
- o HIGHER EXCITED STATES HAVE MORE LOSS CHANNELS
- o METASTABLE TRANSFER BRANCHING
- o EXISTENCE OF SUITABLE ACCEPTOR/LASER SPECIES

Table IV. Identified Critical Issues in SWCL Development.

V = 300 m/sec
H = 10 cm
L = 100 cm
$h\nu$ = 2 ev

DENSITY	POWER
10^{17} cm^{-3}	1000 kW
10^{16}	100
10^{15}	10

Table III. Power Contained in Excited Flow Through Supersonic Nozzle Exit Plane

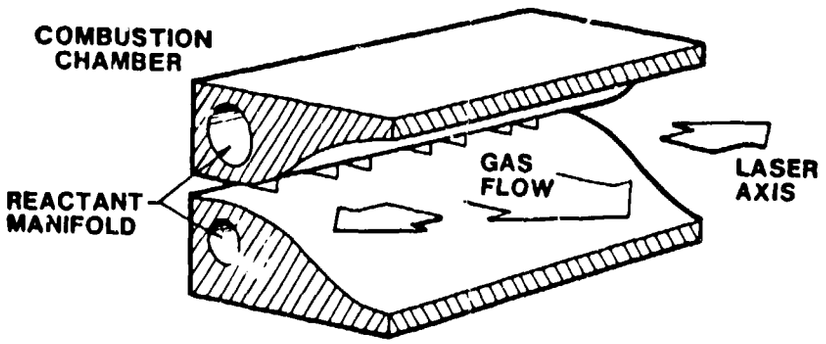

Fig. 1. Schematic of Simplified Supersonic Flow Nozzle for Chemical Lasers

PARAMETER	REQUIREMENT	MOTIVATION
o Laser Wavelength	λ = uv - visible	Brightness
o Transition Energy	E = 2 - 4 ev	Direct
o Radiative Lifetime	$\tau_R \sim 10\ \mu s$	Mixing
o Power in Flow	$\delta \geq 100\ W/cm^2$	Volume
o Fuel Efficiency	$\sigma \geq 1\ MJ/kg$	Weight
o Reactant Mass	$m_r \leq 300$ amu	Weight

Table II. Guidelines for Key SWCL Parameters

- Direct

  $A + BC \rightarrow \underline{AB}^* + C$

- Indirect

  $A + BC \rightarrow AB + C^M$

  $C^M + XY \rightarrow C + \underline{XY}^*$

- Energy Pooling

  $A^* + B^* \rightarrow \underline{C}^*$

- Pulsed

  $h\nu_p + ABC \rightarrow$ Chain Reaction

  $\rightarrow AB^*$

Table V. Generic SWCL Reactions